

Form PTO-1390 (Rev. 5-93)		US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NO. <b>H 3544 PCT/US</b>
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (if known, see 37 CFR 2.5) <b>09/857638</b>
INTERNATIONAL APPLICATION NO. <b>PCT/EP99/09303</b>	INTERNATIONAL FILING DATE <b>November 30, 1999</b>	PRIORITY DATE CLAIMED <b>December 9, 1998</b>	
TITLE OF INVENTION <b>ADHESIVE WITH MAGNETIC NANOPARTICLES</b>			
APPLICANT(S) FOR DO/EO/US <b>Christian KIRSTEN, Hermann ONUSSEIT and Peter CHRISTOPHLIEMK</b>			
Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:			
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)).</p> <p>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p>Items 11. to 16. below concern other document(s) or information included:</p> <p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment</p> <p><input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input type="checkbox"/> Other items or information.:</p>			
"Express Mail" mailing label number <u>EL 615774842 US</u>			

U.S. Application No. (If known see  
CFR 1.30)

09/857638

INTERNATIONAL  
APPLICATION NO.  
PCT/EP99/09303

531 Rec'd PCT

07 JUN 2001

ATTORNEY'S DOCKET NUMBER  
H 3544 PCT/US

17. ■ The following fees are submitted:

**Basic National Fee (37 CFR 1.492(a)(1)-(5)):**

Search Report has been prepared by the EPO or JPO.....\$860.00

International preliminary examination fee paid to USPTO (37CFR 1.482)  
.....\$670.00

No international preliminary examination fee paid to USPTO (37 CFR 1.482)  
but  
international search fee paid to USPTO (37CFR 1.445(a)(2)).....\$690.00

Neither international preliminary examination fee (37CFR 1.482) nor  
international search fee (37 CFR 1.445(a)(2)) paid to USPTO.....\$970.00

International preliminary examination fee paid to USPTO (37CFR 1.482)  
and all claims satisfied provisions of PCT Article 33(2)-(4).....\$96.00

**ENTER APPROPRIATE BASIC FEE AMOUNT**

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\$ 860

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Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30  
months from the earliest claimed priority date 37 (CFR 1.492(e)).

\$ 0

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Claims

Number filed

Number Extra

Rate

Total Claims

14 - 20 =

0

0 X \$18.00

\$ 0

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Independent Claims

3 - 3 =

0

0 X \$80.00

\$ 0

00

Multiple dependent claims (s)(if applicable)

0

+ \$260.00

\$ 0

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**TOTAL OF ABOVE CALCULATIONS**

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\$ 860

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Reduction by ½ for filing by small entity, if applicable. Verified Small Entity statement must also  
be filed. (Note 37 CFR 1.9, 1.27, 1.28).

\$ 0

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**SUBTOTAL**

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\$ 860

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Processing fee of \$130.00 for furnishing the English translation later the ☐ 20 ☐ 30  
months from the earliest claimed priority date (37CFR 1.492(f)).

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**TOTAL NATIONAL FEE**

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\$ 860

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Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be  
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property

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**TOTAL FEES ENCLOSED**

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Amount to be:  
refunded

\$ \_\_\_\_\_

charged

\$860.00

a. ☐ A check in the amount of \$ \_\_\_\_\_ to cover the above fees is enclosed.

b. ☒ Please charge my Deposit Account No. 01-1250 in the amount of \$860.00 to cover the above fees.  
A triplicate copy of this sheet is enclosed. Order No. 01-0444.

c. ☒ The Assistant Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
overpayment to Deposit Account No. 01-1250. A triplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be  
filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO: Henkel Corporation, Law Dept.  
2500 Renaissance Blvd, Suite 200  
Gulph Mills, PA 19406

SIGNATURE

Glenn E. J. Murphy

NAME ATTORNEY FOR APPLICANT  
33,539

REGISTRATION NUMBER

09/857638

531 Rec'd PCT/ 07 JUN 2001

Express Mail  
Label No. EL 615774842 US

PATENT  
Docket H 3544 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Re: PCT/EP99/09303

International Filing Date: November 30, 1999  
Priority Date: December 9, 1998  
Applicant: KIRSTEN, et al.  
Title: ADHESIVE WITH MAGNETIC  
NANOPARTICLES

PRELIMINARY AMENDMENT

Assistant Commissioner of Patents  
Washington, DC 20231

Please enter the amendments below before examining this  
case on the merits:

IN THE SPECIFICATION:

On page 1, insert below the title:

--CROSS-REFERENCE TO RELATED APPLICATIONS

This application is a U.S. National Stage application  
filed under 35 U.S.C. § 371, claiming priority under 35  
U.S.C. §§ 119 and 365 of International Application No.  
PCT/EP99/09303, filed November 30, 1999, in the European  
Patent Office and DE 198 56 819.3, filed on December 9,  
1998, in the German Patent Office.--

On page 1, between lines 25 and 26, insert the heading  
--DESCRIPTION OF THE INVENTION--.

IN THE ABSTRACT:

Please add to the application as a separate page following the claims the abstract appended to this paper.

IN THE CLAIMS:

Please cancel claims 1-11 without prejudice, and add new claims 12-25:

12. An adhesive comprising one or more organic polymers and one or more paramagnetic or ferromagnetic nanoparticles having a particle size of from 1 to 1000 nm.

13. The adhesive of claim 12, comprising 0.1% to 50% by weight of the nanoparticles.

14. The adhesive claim 12, wherein the nanoparticles comprise at least one compound selected from the group consisting of Fe, Co, Ni, Cr, Mo, W, V, Nb, Ta, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, alloys of two or more of said elements, oxides of said elements, ferrites of said elements except iron, and mixtures thereof.

15. The adhesive of claim 12, wherein the nanoparticles comprise magnetite, macchiemite, goethite, or a ferrite of the general formula  $\text{MeOFe}_2\text{O}_3$ , wherein Me represents an element selected from the group consisting of Mn, Co, Ni, Cu, Zn, Mg or Cd, and mixtures thereof.

16. The adhesive of claim 12, in the form of a pressure sensitive adhesive or a contact adhesive.

17. The adhesive of claim 12, in the form of a hot melt adhesive or a dispersion adhesive.

18. The adhesive of claim 12, wherein the one or more organic polymers comprise one or more synthetic organic polymers selected from the group consisting of polyacrylates, polymethacrylates, polyoxy-alkylenes, polyurethanes, polyesters, polystyrene, polyethylene, polyvinyl esters, ethylene-vinyl acetate copolymers, and mixtures thereof.

19. The adhesive of claim 18, wherein the one or more synthetic organic polymers comprise an ethylene-vinyl acetate copolymer or a mixture of two or more such copolymers.

20. The adhesive of claim 12, wherein one or more of the nanoparticles are bonded ionically, coordinatively or covalently to one or more of the organic polymers.

21. A process for preparing an adhesive composition, comprising combining and mixing one or more organic polymers and one or more paramagnetic or ferromagnetic nanoparticles, and optionally one or more solvents or further additives, to form the adhesive composition.

22. A method of temporarily or permanently binding two or more substrates together, comprising the steps of applying to one or more surfaces of the substrates an adhesive comprising one or more paramagnetic or ferromagnetic nanoparticles having a particle size of from 10 to 300 nm and contacting the one or more substrate surfaces with

applied adhesive with one or more other surfaces of the  
substrates to bind the substrates.

23. The adhesive of claim 13, comprising 0.1% to 40% by  
weight of the nanoparticles.

24. The adhesive of claim 23, comprising 0.1% to 30% by  
weight of the nanoparticles.

25. The adhesive of claim 24, comprising 0.1% to 20% by  
weight of the nanoparticles.

#### REMARKS

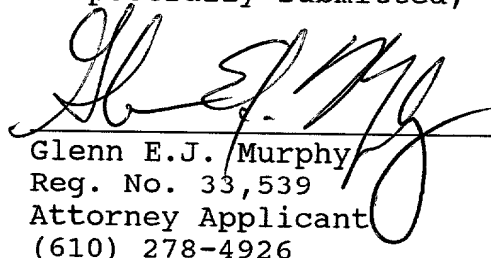
Claims 1-11 have been canceled without prejudice, and  
new claims 12-22 added. The subject matter of the new  
claims is described in the specification at page 3, lines  
27-31, page 4, line 4 to page 5, line 18, page 5, line 35 to  
page 6, line 13, page 9, line 35 to page 10, lines 15, and  
page 10, line 35 to page 11, line 29, as well as in the  
claims as originally filed. The specification has been  
amended to include a cross-reference to related applications  
and headings appropriate to U.S. practice. No new matter  
has been added.

The new claims better claim the full literal and  
equivalent scope and breadth of subject matter disclosed in  
the application, notwithstanding applicants' belief that the  
original claims, drafted for examination in the German and  
European Patent Offices, would have been allowable but for  
minor matters of form permitted in German or European  
practice but objected to in the U.S.P.T.O. The new claims  
find support in the application independent of the original

claims and therefore are not believed to constitute  
narrowing amendments to the original claims within the  
holding of Festo Corp. v. Shoketsu Kinzoku Kogyo Kabushiki  
Co., No. 95-1066 (Fed. Cir. Nov. 29, 2000).

Applicants respectfully request entry of this Amendment  
and examination of the application. If any fees are due to  
enter this paper that have not been accounted for, please  
charge Deposit Account No. 01-1250.

Respectfully submitted,

  
Glenn E.J. Murphy  
Reg. No. 33,539  
Attorney Applicant  
(610) 278-4926

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2500 Renaissance Blvd., Suite 200  
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09/857638  
531 Rec'd PCT/ 07 JUN 2001

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ABSTRACT

An adhesive containing magnetic nanoparticles, a method for producing said adhesive, and the use of an adhesive containing nanoparticles.

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Adhesive with magnetic nanoparticles

The invention relates to an adhesive comprising magnetic nanoparticles, to processes for its preparation, and to the use of an adhesive comprising nanoparticles.

In a society concerned with preserving raw materials, the reuse of materials is playing an ever greater part. The preservation of renewable raw materials is of particular importance. Within that context, a special position is occupied by the pulp producing and pulp processing industry, which is especially reliant on the renewable raw material wood.

In order to preserve wood resources, a procedure adopted ever more frequently and with great success in the past has been to recycle waste paper, which contributes significantly to preserving natural cellulose resources. In recycling, waste paper is generally processed to a pulp, bleached where appropriate, and - alone or together with primary pulp from natural cellulose suppliers - is placed on screens, dried and then processed into rolls. A problem with the recycling of waste paper, however, is that its utilization as pulp often leads to problems during the subsequent processing into paper rolls.

Waste paper commonly includes residues of adhesive originating from paper products made using adhesive (books, brochures, sticky notes, and the like) and from papers bonded on the part of users. Where such residues of adhesive are introduced in comminuted form into the processing operation from pulp to paper rolls, the drying of the substantially dewatered pulp may be accompanied by fusion of the residues of adhesive. Where such dried paper webs with fused adhesive residues are subsequently processed to paper rolls on

papermaking and paper processing machines, the fused adhesive residues may cause the paper web to adhere to the production or processing equipment, or may cause sticking of the paper roll itself. Both phenomena may lead to production stoppages and to reduced product quality. In waste paper processing, therefore, the removal of the residues of adhesive (generally referred to as stickies) is very important.

- 10 US-A 4 176 054 describes a magnetic hot melt adhesive produced by mixing a ground magnetic substance with the hot melt adhesive in liquid state and then using this hot melt adhesive to bond paper. When, in the course of processing, waste paper bonded with such an adhesive is subjected to a magnetic field, the adhesive-containing portions of the waste paper are said to be separable whereas the non-adhesive-containing constituents may be utilized further, as normal, in the waste paper processing operation. To separate the adhesive-containing fractions of the waste paper it is proposed that the waste paper pieces first be comminuted and then, either in the comminuted state or in a mixture with water, be subjected to a magnetic field.
- 20
- 25 A problem with the prior art magnetizing methods, however, is that owing to the use of relatively large magnetic particles it is very difficult to achieve particularly homogeneous and finely divided mixing with adhesives. The result, generally, is an inhomogeneous distribution of the magnetic particles in the adhesive.
- 30

Where waste paper which has been treated with such an adhesive is to be recycled, it is commonly subjected first to a comminuting process. In the course of this process, the adhesives adhering to the waste paper are also comminuted and may be detached from the paper. As the degree of comminution of the adhesive particles increases, however, there is an increase in the likelihood, owing to the inhomogeneous distribution of

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the magnetic particles or because of their size, that individual adhesive particles will no longer contain any magnetic particles. In the magnetic field, such adhesive particles cannot be separated from the remaining waste paper. They are then a cause of the above-described stickies in the production process, which can lead to severe damage and stoppages on the papermaking and paper processing machines.

Often, the intention is not that adhesives should be solid at room temperature in the form of hot melt adhesives; instead, what are desired are liquid adhesives which can be used without high energy consumption on application. Since the solvent based adhesives are unwanted in many countries for ecological reasons, water based dispersion adhesives are a frequently employed alternative here. The use of magnetic particles in dispersion adhesives, however, is difficult, since it is often impossible to incorporate the magnetic particles stably in a dispersion, and phase separation occurs during storage.

It is, accordingly, an object of the present invention to provide an adhesive which does not have the disadvantages of the prior art.

The present invention therefore provides an adhesive comprising at least one polymer and paramagnetic, superparamagnetic or ferromagnetic nanoparticles, or a mixture thereof, having a particle size of from 1 to 1000 nm.

For the purposes of the present invention, an adhesive is a material used for temporary or permanent sticking together of two substrates, especially packaging materials such as paper or cardboard. The term adhesive as used in the context of the present text refers in particular to hot melt adhesives, dispersion adhesives,

pressure sensitive adhesives, hot melt pressure sensitive adhesives, and the like.

For the purposes of the present invention,  
5 nanoparticles are particulate solids which cover a particle size of from about 1 to about 1000 nm, for example from about 2 to about 500 nm, or, for instance, from about 5 to about 300 nm, e.g., about 200 nm or about 30 to about 100 nm. The size figures here refer  
10 to the entirety of the nanoparticles that are present in the adhesive, where at least 90% by weight of the nanoparticles should comply with the abovementioned size figures.

15 The nanoparticles to be used in the context of the present invention have magnetic properties, especially ferromagnetic properties. In one preferred embodiment of the invention, therefore, the nanoparticles comprise at least one element selected from the group consisting  
20 of Fe, Co, Ni, Cr, Mo, W, V, Nb, Ta, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, alloys of two or more of said elements, oxides of said elements or ferrites of said elements, or a mixture of two or more thereof.

25 For example, the nanoparticles may comprise magnetite, macchiemite, goethite or ferrite of the general formula  $\text{MeOFe}_2\text{O}_3$ , Me representing an element selected from the group consisting of Mn, Co, Ni, Cu, Zn, Mg or Cd, or a  
30 mixture of two or more thereof.

Also suitable for use as nanoparticles in the context of the present invention are materials such as wolframite ( $\text{FeMnWO}_4$ ), ferberite ( $\text{FeWO}_4$ ), permanent-  
35 magnetic aluminum nickel cobalt alloys, whose principal constituents comprise iron, cobalt, nickel, aluminum, copper or titanium or mixtures of two or more thereof. Also suitable are alloys of platinum and cobalt, alloys of iron, cobalt, vanadium and chromium, ludwigite

( $\text{Mg}_2\text{Fe}^{3+}[\text{O}_2/\text{BO}_3]$ ), vonsenite ( $\text{Fe}_2^{2+}\text{Fe}^{3+}[\text{O}_2/\text{BO}_3]$ ), cobalt nickel gravels of the general formula  $\text{A}^{2+}\text{B}^{3+}_2\text{X}^{2-}_4$  in which A stands for iron, cobalt, nickel or copper, B stands for iron, cobalt, nickel or chromium or a mixture of two or more thereof, and X stands for S, Se or Te or a mixture of two or more thereof, iron oxides such as iron(II) oxide ( $\text{FeO}$ ) or iron(III) oxide ( $\text{Fe}_2\text{O}_3$ ) in its ferromagnetic modification,  $-\text{Fe}_2\text{O}_3$  (macchiemite) with spinel, magnetite ( $\text{Fe}_3\text{O}_4$ ), cobalt alloys such as the alloys with a Co-Cr matrix commonly used as high temperature matierals, Ni-Fe-Al-Co casting alloys containing up to about 36% by weight cobalt, alloys of the type CoCrW, chromium(IV) oxide ( $\text{CrO}_2$ ), the oxide ceramic materials for classification in the group of the ferrites, of the general composition  $\text{M}^2\text{Fe}^3_2\text{O}_4$  or  $\text{M}^2\text{O}^*\text{Fe}_2\text{O}_3$ , which contain permanent magnetic dipoles, M representing zinc, cadmium, cobalt, manganese, iron, copper, magnesium and the like, and also iron itself.

The preparation of magnetite or macchiemite nanoparticles may be achieved, for example, by using a microemulsion technology. Here, the disperse phase of a microemulsion is used to limit the size of the particles that are formed. In a W/O microemulsion, a metal containing reagent is dissolved in the disperse aqueous phase. The reagent is subsequently reacted in the disperse phase to give a precursor of the desired magnetic compound, which thereafter already has the desired size in the nanometer range. Subsequently, with a careful oxidation step, the metal oxide, especially iron oxide in the form of magnetite or macchiemite, is prepared. An appropriate process is described, for example, in US-A 5 695 901.

Adhesives suitable in the context of the present invention include in particular those adhesives as used to bond paper. Such adhesives commonly include at least one organic synthetic polymer or a natural organic polymer as occurs in nature or may be recovered from

natural substances. Likewise suitable in the context of the present invention are adhesives comprising a mixture of one or more organic synthetic polymers and one or more organic polymers.

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The organic synthetic polymers as used in one preferred embodiment in the context of the present invention include, for example, polyesters, polyethers, polyamides, polyurethanes, polyacrylates, polymeth-  
10 acrylates, polyvinyl acetate, ethylene, vinyl acetate copolymers, propylene-vinyl acetate copolymers, styrene-acrylate and styrene-methacrylate copolymers, and the like.

15 The adhesive of the invention with magnetic nanoparticles may be a hot melt adhesive or a dispersion adhesive in an embodiment as a pressure sensitive adhesive or as a contact adhesive.

20 The adhesive of the invention may be used, for example, as a hot melt adhesive. For the purposes of the present invention, hot melt adhesives are adhesives which at room temperature are solid and are also at least substantially water- and solvent-free. Hot melt  
25 adhesives are applied from the melt to plies of paper that are to be bonded, and on cooling they set physically and solidify. Examples of suitable hot melt adhesives include organic polymers such as polyesters, polyurethanes, polyamides, polyalkylene oxides or  
30 addition polymers, examples being ethylene-vinyl acetate copolymers, or mixtures of two or more of said polymers, or formulations comprising one of said polymers or a mixture of two or more thereof.

35 Polyurethanes, for example, may be used as hot melt adhesive in the context of the present invention.

Polyurethanes as may be used as hot melt adhesive in the context of the present invention are commonly

prepared by reacting at least one polyisocyanate, preferably a diisocyanate, and a polyol component consisting preferably predominantly of diols. The polyol component may contain only one polyol; alternatively, a mixture of two or more different polyols may be used as the polyol component. Polyalkylene oxides, for example, are suitable as the polyol component or at least as part of the polyol component.

10

If desired, parts of the polyalkylene oxide may be replaced by other hydrophobic diols containing ether groups and having molecular weights of from 250 to 3000, preferably from 300 to 2000, in particular from 500 to 1000. Specific examples of such diols are polypropylene glycol (PPG), polybutylene glycol, polytetrahydrofuran, polybutadienediol and alkanediols having from 4 to 44 carbon atoms. Preferred hydrophobic diols are polypropylene glycol, polytetrahydrofuran having a molecular weight of from 500 to 1000, and also 1,10-decanediol, 1,12-dodecanediol, 1,12-octadecanediol, dimer fatty acid diol, 1,2-octanediol, 1,2-dodecanediol, 1,2-hexadecanediol, 1,2-octadecanediol, 1,2-tetradecanediol, 2-butene-1,4-diol, 2-butyne-1,4-diol, 2,4,7,9-tetramethyl-5-decyne-4,7-diol and also its ethoxylation products, especially with up to 30 mol of ethylene oxide.

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Besides the diols of the polyol component, diisocyanates are key building blocks of the polyurethane which can be used as hot melt adhesive. These are compounds of the general structure  $O=C=N-X-N=C=O$ , X being an aliphatic, alicyclic or aromatic radical, preferably an aliphatic or alicyclic radical having from 4 to 18 carbon atoms.

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Examples that may be mentioned of suitable isocyanates include 1,5-naphthylene diisocyanate, 4,4'-diphenylmethane diisocyanate (MDI), hydrogenated MDI ( $H_{12}$ MDI),

xylylene diisocyanate (XDI), tetramethylxylylene diisocyanate (TMXDI), 4,4'-diphenyldimethylmethane diisocyanate, di- and tetraalkylenediphenylmethane diisocyanate, 4,4'-dibenzyl diisocyanate, 1,3-phenylene diisocyanate, 1,4-phenylene diisocyanate, the isomers of tolylene diisocyanate (TDI), 1-methyl-2,4-diisocyanatocyclohexane, 1,6-diisocyanato-2,2,4-trimethylhexane, 1,6-diisocyanato-2,4,4-trimethylhexane, 1-isocyanatomethyl-3-isocyanato-1,5,5,-trimethylcyclohexane (IPDI), chlorinated and brominated diisocyanates, phosphorus diisocyanates, 4,4'-diisocyanatophenylperfluoroethane, tetramethoxybutane 1,4-diisocyanate, butane 1,4-diisocyanate, hexane 1,6-diisocyanate (HDI), dicyclohexylmethane diisocyanate, cyclohexane 1,4-diisocyanate, ethylene diisocyanate, bisisocyanatoethyl phthalate, and also diisocyanates containing reactive halogen atoms, such as 1-chloromethylphenyl 2,4-diisocyanate, 1-bromomethylphenyl 2,6-diisocyanate, 3,3-bis-chloromethyl ether 4,4'-diphenyl diisocyanate.

Sulfur polyisocyanates are obtained, for example, by reacting 2 mol of hexamethylene diisocyanate with 1 mol of thiodiglycol or dihydroxydihexyl sulfide. Further diisocyanates that may be used are, for example, trimethylhexamethylene diisocyanate, 1,4-diisocyanatobutane, 1,12-diisocyanatododecane, and dimer fatty acid diisocyanate. The following are especially suitable: tetramethylene, hexamethylene, undecane, dodecamethylene, 2,2,4-trimethylhexane, 1,3-cyclohexane, 1,4-cyclohexane, 1,3- and/or 1,4-tetramethylxylylene, isophorone, 4,4-dicyclohexylmethane, and lysine ester diisocyanate. Very particular preference is given to tetramethylxylylene diisocyanate (TMXDI), especially the m-TMXDI from Cyanamid.

To further increase the molecular weight it is possible, for example, to perform a chain extension in a known manner. For this purpose, prepolymers are prepared first of all using excess diisocyanate, and



are then extended using short-chain amino alcohols, diols or diamines or using water, with an increase in the molecular weight.

5 Preferably, however, the polyurethane is prepared in a single stage process. In this case, for example, first all of the starting materials are mixed in the presence of an organic solvent, with a water content of less than 0.5% by weight. The mixture is heated at from 80  
10 to 200°C, in particular at from 100 to 180°C and preferably at from 130 to 170°C, for from about 1 to 30 hours. The reaction time may be shortened by the presence of catalysts. In particular it is possible to use tertiary amines, e.g., triethylamine,  
15 dimethylbenzylamine, bisdimethylaminoethyl ether and bismethylaminomethylphenol. Particularly suitable are 1-methylimidazole, 2-methyl-1-vinylimidazole, 1-allyl-imidazole, 1-phenylimidazole, 1,2,4,5-tetramethylimidazole, 1-(3-aminopropyl)imidazole, pyrimidazole, 4-di-  
20 methylaminopyridine, 4-pyrrolidinopyridine, 4-morpholinopyridine or 4-methylpyridine. It is preferred, however, to work without a catalyst. The solvent as well is appropriately omitted. By solvents in the context of the present text are meant inert organic  
25 liquid substances having a boiling point of less than 200°C under atmospheric pressure.

If the adhesive of the invention is to be a dispersion adhesive, then in one preferred embodiment it comprises  
30 an organic synthetic polymer selected from the group consisting of polyacrylates, polymethacrylates, polystyrene, polyvinyl esters, ethylene-vinyl acetate copolymers or acrylate-styrene copolymers.

35 In another preferred embodiment of the invention, the adhesive of the invention is a dispersion adhesive.

Instead or in addition to one or more organic synthetic polymers the adhesive of the invention may comprise an

organic natural polymer or a mixture of two or more thereof. By an organic natural polymer are meant polymers as may be obtained from natural substances by simple chemical operations. In the context of the present invention, the term also includes simple derivatives of organic natural polymers, examples being the esterification derivatives or alkoxylation derivatives of starch or of cellulose.

10 In one preferred embodiment of the present invention, the adhesive of the invention contains the nanoparticles in an amount of from about 0.1 to about 50% by weight, preferably in an amount of from about 1 to about 30% by weight, in particular from about 2 to about 20% by weight.

The adhesive of the invention contains the organic synthetic polymer or the organic natural polymer or the mixture of one or more organic synthetic polymers and one or more organic natural polymers in an amount of at least about 10% by weight. If the adhesive of the invention is to be used as a hot melt adhesive, then it is advantageous if it contains at least one synthetic organic polymer or one natural organic polymer in a relatively large amount, for example, at least about 50% by weight.

Likewise suitable as adhesives in the context of the present invention are hot melt adhesives containing postcrosslinking groups, as are commonly used to produce bonds possessing particular heat stability. In this context, the use of polyurethanes as synthetic organic polymer is particularly appropriate.

35 The adhesive of the invention may further comprise a heat sealing adhesive. By heat sealing adhesives are meant heat activatable adhesives which are applied as a solution, emulsion, dispersion or melt to the surface of the substrates to be sealed. There, initially owing

to the evaporation of the solvents or by cooling, they set to give a nontacky adhesive film. The subsequent bonding of the substrates generally takes place, following their joining and pressing together, by heating in hot presses or in a high frequency field. On cooling, the workpieces are bonded with solidification of the heat sealing adhesive film. Especially suitable for use in heat sealing adhesives are, for example, copolymers based on ethylene, (meth)acrylates, vinyl chloride, vinylidene chloride, vinyl acetate, and also polyamides, polyesters and polyurethanes.

The adhesive of the invention may further comprise a pressure sensitive adhesive. Pressure sensitive adhesives are generally viscoelastic adhesives which in solvent-free form at 20°C are permanently tacky and remain adherable, and, with low substrate specificity, adhere immediately to virtually all substrates under gentle applied pressure. Bonds produced using pressure sensitive adhesives can usually be parted without destroying the bonded substrates. In the context of the present invention, pressure sensitive adhesives comprise as organic synthetic polymer, for example, natural and synthetic rubbers, polyacrylates, polyesters, polychloroprenes, polyisobutenes, polyvinyl ethers and polyurethanes. If desired, the pressure sensitive adhesives may further comprise additives which promote, for example, one-sided redetachability from paper surfaces.

In another preferred embodiment of the invention, the adhesive of the invention is a dispersion adhesive. Adhesives termed dispersion adhesives are mostly aqueous dispersions of organic polymers which are capable of bonding wood, paper, cardboard, wall coverings, leather, felt, cork, textiles, plastics or metals. Dispersion adhesives set by evaporation of the dispersion medium (water) to form an adhesive film. Examples of suitable synthetic organic polymers in

dispersion adhesives are polyacrylates, polymethacrylates, polyurethanes, polyesters, polyvinyl acetals, ethylene-vinyl acetate (EVA) copolymers, and the like.

5

In addition to the abovementioned organic synthetic or natural polymers, the adhesive of the invention may comprise further additives which influence, for example, the sticking properties, the aging behavior, the setting process, or the adhesion. For example, the adhesive may include what are known as tackifier resins, which can generally be subdivided into natural and synthetic (synthetic resins). They include, for example, alkyd resins, epoxy resins, melamine resins, phenolic resins, urethane resins, hydrocarbon resins, and also natural resins such as rosin, wood terpentine oil and tall oil. The synthetic resins include hydrocarbon resins, ketone resins, coumarone-indene resins, isocyanate resins, and terpene phenolic resins.

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Furthermore, adhesives of the invention may comprise solvents. Examples of suitable solvents are mono or polyhydric alcohols having from about 2 to about 10 carbon atoms.

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Additionally, the adhesives of the invention may comprise defoamers. Examples of suitable defoamers are fatty alcohol based or silicone based defoamers.

30 The adhesives may additionally comprise protective colloids such as polyvinylpyrrolidones, polyvinyl alcohols, cellulose or cellulose derivatives.

Additives which the adhesive of the invention may further comprise include stabilizers or antioxidants. Included among these, generally, are phenols, sterically hindered phenols of high molecular weight, polyfunctional phenols, sulfur and phosphorus containing phenols or amines. Examples of suitable

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stabilizers include hydroquinone, hydroquinone methyl ether 2,3-(di-tert-butyl)hydroquinone, 1,3,5-trimethyl-2,4,6-tris (3,5-di-tert-butyl-4-hydroxybenzyl) benzene; pentaerythritol tetrakis-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate; n-octadecyl 3,5-di-tert-butyl-4-hydroxyphenyl)propionate; 4,4-methylenebis(2,6-di-tert-butylphenol); 4,4-thiobis(6-tert-butyl-o-cresol); 2,6-di-tert-butylphenol; 6-(4-hydroxyphenoxy)-2,4-bis-(n-octylthio)-1,3,5-triazine; di-n-octadecyl 3,5-di-tert-butyl-4-hydroxybenzylphosphonates; 2-(n-octylthio)-ethyl-3,5-di-tert-butyl-4-hydroxybenzoate; and sorbitol hexa[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate]; and also p-hydroxydiphenylamine or N,N'-diphenylenediamine or phenothiazine.

The adhesive of the invention may further comprise plasticizers such as benzoate plasticizers, phosphate plasticizers, liquid resin derivatives or vegetable and animal oils. Suitable examples include sucrose benzoate, diethylene glycol dibenzoate and/or diethylene glycol benzoate in which from about 50 to about 95% of all hydroxyl groups have been esterified, phosphate plasticizers, for example, t-butylphenyl diphenyl phosphate, polyethylene glycols and their derivatives, for example, diphenyl ethers of poly(ethylene glycol), liquid resin derivatives, for example, the methyl esters of hydrogenated resin, vegetable and animal oils, for example, glycerol esters of fatty acids and their polymerization products.

Likewise suitable are plasticizers based on phthalic acid, especially the alkyl phthalates.

The adhesive of the invention may further comprise dyes such as titanium dioxide, fillers such as talc, clay and the like, and also pigments.

Where the adhesive of the invention comprises an adhesive which is postcrosslinking, for example,

through the influence of electron beams or UV rays it is possible for photoinitiators to be present as additional additives in the adhesive. Said photoinitiators may, for example, comprise Norrish type

5 I fragmenting substances such as benzophenone, hydroquinone, photoinitiators of the Irgacure<sup>®</sup>, Darocure<sup>®</sup> or Speedcure<sup>®</sup> range (manufacturer: Ciba-Geigy). If desired, the adhesive of the invention may comprise a monofunctional reactive diluent which is

10 polymerizable, for example, by irradiation with UV light or with electron beams. Particularly suitable for this purpose are the corresponding esters of acrylic acid or methacrylic acid. Example of such esters include N-butyl acrylate, 2-ethylhexyl acrylate,

15 3-methoxybutyl acrylate, 2-phenoxyethyl acrylate, benzyl acrylate or 2-methoxypropyl acrylate.

The adhesives of the invention may further comprise emulsifiers or stabilizers or a mixture thereof.

20 Suitable emulsifiers are generally surfactants which have a hydrophilic and a hydrophobic group. These may be anionic emulsifiers, cationic emulsifiers or amphoteric emulsifiers. Suitable examples include hydrocarbon emulsifiers containing from about 6 to

25 about 22 carbon atoms, the hydrocarbon chain being branched, unbranched, saturated, unsaturated, substituted, aliphatic or aromatic.

For the preparation of the adhesives of the invention,

30 the organic synthetic polymer or the organic natural polymer or the mixture of one or more organic synthetic polymers and one or more organic natural polymers are mixed with the nanoparticles and, if desired, with a solvent and further additives. If the adhesive of the

35 invention is to be a hot melt adhesive, then mixing may take place in the melt of the hot melt adhesive; however, it is likewise possible to add the nanoparticles as early as during the preparation of the polymer which is used as the hot melt adhesive.

Where the adhesive of the invention is to be a dispersion adhesive, the nanoparticles may be incorporated directly into the polymer dispersion of  
5 the dispersion adhesive.

In another preferred embodiment of the invention, the nanoparticles are added to a dispersion adhesive of the invention even before the preparation of the synthetic  
10 organic polymer. In this case the dispersion adhesive of the invention is prepared by means of an emulsion polymerization in the course of which, customarily, droplets of monomers needed to prepare the subsequent polymer are polymerized in an aqueous emulsion. The  
15 nanoparticles may be added to the emulsion even before the polymerization, leading to a particularly homogeneous distribution of the nanoparticles in the dispersion adhesive.

20 In a further preferred embodiment of the present invention, the nanoparticles are directly attached ionically, coordinatively or covalently to an organic synthetic or organic natural polymer.

25 For this purpose the nanoparticles are surface modified in such a way that, subsequently, a reaction with a functional group of an organic natural or synthetic polymer is possible or it is possible for the modified nanoparticle to be incorporated into an organic  
30 synthetic polymer.

In one preferred embodiment of the invention, the surface of the nanoparticles is modified, for example, with silanes. Where oxides are used as nanoparticles,  
35 these oxides generally carry surface OH groups which are able to react with silanes or halosilanes to form a covalent Si-O bond. If the silanes, for their part, possess an appropriate functional group which permits subsequent attachment of the silanes to a polymer, this

may be used to affix the modified nanoparticles covalently to the polymer. Suitable functional groups are, for example, olefinically unsaturated double bonds or protected OH or NH groups. An appropriate  
5 possibility for modifying the surface of nanoparticles with silane compounds is described, for example, in US-A 5,695,901.

The invention additionally provides a process for  
10 preparing an adhesive, characterized in that a polymer, paramagnetic or ferromagnetic nanoparticles or a mixture thereof and, where appropriate, solvents or further additives or a mixture of two or more thereof are mixed.

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The present invention likewise provides for the use of paramagnetic or ferromagnetic nanoparticles or of a mixture of one or more paramagnetic and one or more ferromagnetic nanoparticles having a particle size of  
20 from about 10 to about 300 nm in adhesives.



**Claims**

1. An adhesive comprising at least one organic polymer or a mixture of two or more organic polymers and paramagnetic or ferromagnetic nanoparticles having a particle size of from 1 to 1000 nm, or a mixture thereof.
2. The adhesive as claimed in claim 1, characterized in that it contains the nanoparticles in an amount of from 0.1 to 40% by weight.
3. The adhesive as claimed in one of claims 1 or 2, characterized in that the nanoparticles comprise at least one compound selected from the group consisting of Fe, Co, Ni, Cr, Mo, W, V, Nb, Ta, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, alloys of two or more of said elements, oxides of said elements or ferrites of said elements (besides iron), or a mixture of two or more thereof.
4. The adhesive as claimed in one of claims 1 to 3, characterized in that the nanoparticles comprise magnetite, macchiemite, goethite or a ferrite of the general formula  $\text{MeOFe}_2\text{O}_3$ , Me representing an element selected from the group consisting of Mn, Co, Ni, Cu, Zn, Mg or Cd, or a mixture of two or more thereof.
5. The adhesive as claimed in one of claims 1 to 4, characterized in that it is a pressure sensitive adhesive or a contact adhesive.
6. The adhesive as claimed in one of claims 1 to 5, characterized in that it is a hot melt adhesive or a dispersion adhesive.

7. An adhesive, characterized in that there is present as synthetic organic polymer a polymer selected from the group consisting of polyacrylates, polymethacrylates, polyoxy-alkylenes, polyurethanes, polyesters, polystyrene, polyethylene, polyvinyl esters, ethylene-vinyl acetate copolymers; or a mixture of two or more thereof.
8. The adhesive as claimed in one of claims 1 to 7, characterized in that it comprises as synthetic organic polymer an ethylene-vinyl acetate copolymer or a mixture of two or more such copolymers.
9. The adhesive as claimed in one of claims 1 to 8, characterized in that the nanoparticles are bonded ionically, coordinatively or covalently to the organic polymer.
10. A process for preparing an adhesive, characterized in that a synthetic organic polymer, paramagnetic or ferromagnetic nanoparticles, or a mixture thereof and, where appropriate, solvents or further additives, or a mixture of two or more thereof, are mixed.
11. The use of paramagnetic or ferromagnetic nanoparticles or of a mixture of one or more paramagnetic and one or more ferromagnetic nanoparticles having a particle size of from 10 to 300 nm in adhesives.

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0010/PTO  
Rev. 6/95

U.S. Department of Commerce  
Patent and Trademark Office

# DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number

H 3544 PCT/US

First Named Inventor

KIRSTEN, Christian

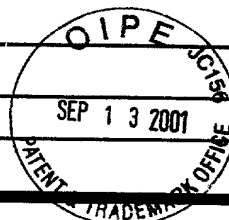
COMPLETE IF KNOWN

Application Number

Filing Date

Group Art Unit

Examiner Name



As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**ADHESIVE WITH MAGNETIC NANOPARTICLES**

the specification of which

(Title of the invention)

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY)

11/30/1999

as United States Application Number or PCT International

Application Number

PCT/EP99/09303

and was amended on (MM/DD/YYYY)

(if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)

Country

Foreign Filing Date (MM/DD/YYYY)

Priority Not Claimed

Certified Copy Attached?  
YES NO

198 56 819.3

Germany

12/09/1998

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Application Number(s)

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## DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP99/09303	11/30/1999	

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As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name  Customer Number  or label

☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
Wayne C. Jaeschke	21,062	Glenn E. J. Murphy	33,539
Kimberly R. Hild	39,224	Stephen D. Harper	33,243

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor:

☐ A petition has been filed for this unsigned

Given Name	Christian	Middle Initial		Family Name	KIRSTEN	Suffix e.g. Jr.	
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Inventor's Signature		Date	22.06.01
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City	40789 Monheim	State		Zip		Country	Germany	Applicant Authority	
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☒ Additional inventors are being named on supplemental sheet(s) attached hereto

<b>DECLARATION</b>										<b>ADDITIONAL INVENTOR(S) Supplemental Sheet</b>											
Name of Additional Joint Inventor, if any:						<input type="checkbox"/> A petition has been filed for this unsigned inventor															
Given Name		Hermann				Middle Initial				Family Name		ONUSSEIT		Suffix e.g. Jr.							
Inventor's Signature										Date											
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Post Office Address		Elsa-Brandstroem-Strasse 17																			
Post Office Address																					
City		42781 Haan				State				Zip				Country		Germany		Applicant Authority			
Name of Additional Joint Inventor, if any:						<input type="checkbox"/> A petition has been filed for this unsigned inventor															
Given Name		Peter				Middle Initial				Family Name		CHRISTOPHLIEMK		Suffix e.g. Jr.							
Inventor's Signature										Date											
Residence: City		Duesseldorf				State				Country		Germany		Citizenship		Germany					
Post Office Address		Rudolf-Breitscheid-Strasse 61																			
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Number

H 3544 PCT/US

First Named  
Inventor

KIRSTEN, Christian

COMPLETE IF KNOWN

Application Number

Filing Date

Group Art Unit

Examiner Name

**DECLARATION FOR  
UTILITY OR DESIGN  
PATENT APPLICATION**

☐ Declaration  
Submitted  
with Initial Filing OR ☒ Declaration  
Submitted after  
Initial Filing

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**ADHESIVE WITH MAGNETIC NANOPARTICLES**

the specification of which

(Title of the invention)

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 11/30/1999 as United States Application Number or PCT International

Application Number PCT/EP99/09303 and was amended on (MM/DD/YYYY) (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES NO
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Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.
		<input type="checkbox"/>

Burden Hour Statement: This form is estimated to take .4 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

# DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP99/09303	11/30/1999	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name  Customer Number  or label

OR

☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
Wayne C. Jaeschke	21,062	Glenn E. J. Murphy	33,539
Kimberly R. Hild	39,224	Stephen D. Harper	33,243

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number or label 00423 OR ☐ Fill in correspondence address below

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unsigned

Given Name	Christian	Middle Initial		Family Name	KIRSTEN	Suffix e.g. Jr.	
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Inventor's Signature		Date	
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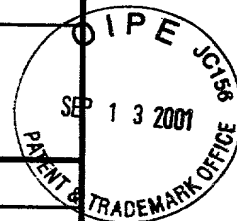
Residence: City	Monheim	State		Country	Germany	Citizenship	Germany
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Post Office Address	Innsbrucker Strasse 23
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Post Office Address	
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City	40789 Monheim	State		Zip		Country	Germany	Applicant Authority	
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☒ Additional inventors are being named on supplemental sheet(s) attached hereto



## DECLARATION

ADDITIONAL INVENTOR(S)  
Supplemental Sheet

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name	Hermann	Middle Initial	Family Name	ONUSSEIT	Suffix e.g. Jr.
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Inventor's Signature	<i>[Signature]</i>	Date	July 11, 2001
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Residence: City	Haan	State	DEX	Country	Germany	Citizenship	Germany
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Post Office Address

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Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name	Peter	Middle Initial	Family Name	CHRISTOPHLEMK	Suffix e.g. Jr.
------------	-------	----------------	-------------	---------------	-----------------

Inventor's Signature	<i>[Signature]</i>	Date	July 11, 2001
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Post Office Address	Rudolf-Breitscheid-Strasse 61
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Post Office Address

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Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial	Family Name		Suffix e.g. Jr.
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Inventor's Signature		Date	
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Residence: City		State		Country		Citizenship	
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Post Office Address

Post Office Address

City		State		Zip		Country		Applicant Authority
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Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial	Family Name		Suffix e.g. Jr.
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Inventor's Signature		Date	
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Residence: City		State		Country		Citizenship	
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Post Office Address

Post Office Address

City		State		Zip		Country		Applicant Authority
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☐ Additional inventors are being named on supplemental sheet(s) attached hereto